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Life cycle assessment of soil and groundwater remediation technologies: literature review

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Abstract
Background, aim, and scope Life cycle assessment (LCA) is becoming an increasingly widespread tool in support systems for environmental decision-making regarding the cleanup of contaminated sites. In this study, the use of LCA to compare the environmental impacts of different remediation technologies was reviewed. Remediation of a contaminated site reduces a local environmental problem, but at the same time, the remediation activities may cause negative environmental impacts on the local, regional, and global scale. LCA can be used to evaluate the inherent trade-off and to compare remediation scenarios in terms of their associated environmental burden.
Main features An overview of the assessed remediation technologies and contaminant types covered in the literature is presented. The LCA methodologies of the 12 reviewed studies were compared and discussed with special focus on their goal and scope definition and the applied impact assessment. The studies differ in their basic approach since some are prospective with focus on decision-support while others are retrospective aiming at a more detailed assessment of a completed remediation project.
Literature review The literature review showed that only few life cycle assessments have been conducted for in situ remediation technologies aimed at groundwater-threatening contaminants and that the majority of the existing literature focuses on ex situ remediation of contaminated soil. The functional unit applied in the studies is generally based on the volume of contaminated soil (or groundwater) to be treated; this is in four of the studies combined with a cleanup target for the remediation. While earlier studies often used more simplified impact assessment models, the more recent studies based their impact assessment on established methodologies covering the conventional set of impact categories. Ecotoxicity and human toxicity are the impact categories varying the most between these methodologies. Many of the reviewed studies address the importance of evaluating both primary and secondary impacts of site remediation. Primary impacts cover the local impacts related to residual contamination left in the subsurface during and after remediation and will vary between different remediation technologies due to different cleanup efficiencies and cleanup times. Secondary impacts are resource use and emissions arising in other stages of the life cycle of the remediation project.
Discussion Among the reviewed literature, different approaches for modeling the long-term primary impacts of site contamination have been used. These include steady state models as well as dynamic models. Primary impacts are not solely a soil contamination or surface water issue, since many frequently occurring contaminants, such as chlorinated solvents, have the potential to migrate to the groundwater as well as evaporate to ambient air causing indoor climate problems. Impacts in the groundwater compartment are not included in established impact assessment methodologies; thus, the potential groundwater contamination impacts from residual contamination are

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difficult to address in LCA of site remediation. Due to the strong dependence on local conditions (sensitivity of groundwater aquifer, use for drinking water supply, etc.) a more site-specific impact assessment approach than what is normally applied in LCA is of relevance.

Conclusions, recommendations, and perspectives The inclusion of groundwater impacts from soil contaminants requires the definition of an impact category covering human toxicity via groundwater or the inclusion of these impacts in the human toxicity impact category and the associated characterization models and normalization procedures. When evaluating groundwater impacts, attention should also be paid to potentially degradable contaminants forming metabolites of higher human toxic concern than the parent compound.

Keywords Ex situ remediation · Groundwater contamination · In situ remediation · Life cycle assessment · Metabolites · Normalization · Primary impacts · Secondary impacts · Soil contamination

1 Background, aim, and scope

The choice of a remedial strategy for a contaminated site entails economical consequences for the decision maker as well as environmental consequences for the local, regional, and global environment. Increasingly, the attention of decision makers, such as regulatory bodies, is paid to the external impacts arising from remediation in order to include this aspect in the technology selection process.

Life cycle assessment (LCA) is a widely accepted and applied method for evaluating and quantifying the environmental impacts associated with the life of a product from extraction of resources to production, use, and end-of-life treatment. Life cycle assessment takes a systems perspective on the product and therefore, lends itself to being applied to the assessment of systems that deliver a function or a service, e.g., a remediation technology (Wenzel et al. 1997; Hauschild 2005).

Duration and efficiency of site remediation depend on the technological approach taken. Thermal methods provide a fast but energy intensive removal of contaminants, whereas biological methods or pump-and-treat methods are less energy intensive, but require much longer operation periods. The systems perspective and the broad coverage of environmental impacts applied in life cycle assessment make it a well-suited tool to support decision-making regulatory bodies and to bring focus to those processes that contribute most to the overall environmental burden of the remediation project.

Despite the standardization of LCA by the International Standards Organization (ISO 14040–14044), many aspects of the analysis rely on choices made by the analyzer. This is especially the case for the impact assessment part, where many models are available. Thus, although a number of LCA applications have been done in the field of soil and groundwater remediation technologies, the results and conclusions drawn are not necessarily comparable.

The objective of this paper is to review the existing literature and compare the methodological approaches previously used for conducting life cycle assessments of remediation projects for contaminated sites. In the discussion, we pinpoint future needs and developments for using LCA in this field. Particularly, we discuss improvements and adjustments of the LCA methodology for sites contaminated with pollutants commonly found in groundwater such as chlorinated solvents. The specific focus on chlorinated solvents is motivated by the fact that trichloroethylene was the most prevalent contaminant and occurred at 50% of the sites prioritized for remediation in the National Priority List by US EPA (1997). Moreover, the applicability of approaches from related environmental fields for including impacts in the groundwater compartment is discussed. Thus, compared to a previous review by Suer et al. (2004), this review both covers more recent publications in the field and has a broader scope. Restoration of brownfields and nature areas (e.g., lakes and rivers) lies outside the scope of the paper.

Soil and groundwater remediation technologies represent a broad spectrum of technological approaches for cleanup of contaminated sites. The aim of the remedial effort can be defined as to reduce, e.g., mass, flux, toxicity, volume, or concentration of the contamination in order to comply with regulative standards or a site-specific risk-based cleanup target.

Overall, remediation technologies can be divided into two groups based on the physical location of the remedial action:

- In ex situ remediation, the contaminated media is removed from the site for subsequent treatment. This can either take place in an above ground treatment facility (on-site) or by treatment or disposal elsewhere (off-site).
- In in situ remediation, the treatment of the contaminated media takes place by actions that target the contamination in its actual location in the subsurface.

The applied remediation technology can either be directed at the "source zone" or the "contaminant plume" or it can be a combination of these. While source zone remediation technologies are often more short-term aggressive technologies, plume zone technologies are typically long-term techniques aiming at control or at treating the remaining contamination in the groundwater after the source zone has been remediated. The number of potentially applicable techniques for a given contaminated site depends on the type

of contaminant and the local hydrogeological setting. Some contaminants, such as many metals and polyaromatic hydrocarbons (PAHs), have a high tendency for sorption to the soil particles and are less frequently found in deeper soil layers and groundwater aquifers (Fetter 1999; Mackay et al. 1992). Other contaminants are very mobile in the subsurface and are frequent groundwater contaminants. Chlorinated aliphatic hydrocarbons such as tetrachloroethylene (PCE) and trichloroethylene (TCE) are among the most frequently observed contaminants threatening groundwater quality (Stroo et al. 2003) due to their extensive and widespread use as cleaning agents, solvents, and degreasers. Chlorinated solvents belong to the group of dense nonaqueous phase liquids (DNAPLs). DNAPLs are characterized as liquids being denser than water, immiscible with water, and having a high viscosity. These characteristics enable a spill to migrate as a separate liquid phase through the subsurface leaving behind a trace of residual phase contamination trapped in the soil pores. The separate phase will due to the high density be able to penetrate into deep aquifers. When an impermeable layer is reached, pools of contamination are formed and serve as a long-term source to contamination of the groundwater (Stroo et al. 2003). Due to the low water solubility and the complex spatial distribution of these contaminants in the subsurface, conventional pump-and-treat techniques have shown not to be very effective and to require very extended operation periods (McGuire et al. 2006; Stroo et al. 2003). Innovative in situ remediation technologies for source and plume remediation of chlorinated solvents have thus been developed and applied during the recent years. These technologies cover chemical, biological, and thermal treatment methods, e.g., chemical oxidation, stimulated bioremediation, reduction with zerovalent iron, and remediation assisted by electrical heating or steam injection.

2 Literature review: LCA methodology comparison

Among the studies located by the authors of this paper, Beinat et al. (1997) were the first to apply the principles of LCA to site remediation technologies in their risk reduction, environmental merit, and costs methodology (REC). This was followed by Diamond and colleagues (1999) who proposed a framework for life cycle management of site remediation as well as an impact assessment method for qualitative life cycle assessment. The framework was applied by Page et al. (1999) for a life cycle assessment of treatment of lead contaminated soil. The life cycle assessment studies of site remediation show differences in their fundamental approach and goal. Some are retrospective and aiming at a more detailed assessment of a completed remediation project, whereas others are prospective assessments intended to serve as decision-support in the choice between different remedial options. The latter may have a more strategic perspective and address generic cases based on less site-specific data.

The focus in our comparison of methodologies is on the issues and differences of importance for application of LCA to site remediation, as they are found in the initial goal and scope phase, in particular the definition of functional unit and drawing of system boundaries, and in the impact assessment phase. The reviewed literature covers articles and reports published during the latest 12 years. Table 1 gives an initial overview of remediation technologies (in situ/ex situ) and contaminant types included in the 12 studies. As seen from the table, the majority of the studies focus on ex situ remediation technologies and decontamination of soil, whereas only four studies (Cadotte et al. 2007; Bayer and Finkel 2006; ScanRail Consult et al. 2000; Diamond et al. 1999) cover in situ technologies for groundwater remediation. The in situ remediation technologies covered are limited to soil vapor extraction systems, biosparging, bioremediation, natural attenuation, and chemical oxidation and do not represent the recent advances in the field of in situ remediation of chlorinated solvents source zones and groundwater plumes. This reflects the fact that ex situ remediation has been carried out more frequently than in situ which have been developed through recent years.

2.1 Goal and scope and data inventory

The reviewed studies listed in Table 1 can be divided into two groups based on the primary goal of the LCA: (1) LCA framework studies for prospective analysis to be used as decision-support and (2) in-depth retrospective life cycle assessment of one or more remediation techniques. Two of the studies are suitable for both goals (Diamond et al. 1999; ScanRail Consult et al. 2000). A life cycle assessment compares the environmental impacts associated with providing a defined functional unit, which describes the service that all compared technologies fulfill. A functional unit is explicitly defined in nine out of the 12 reviewed studies (see Table 2). Four of these specify a certain cleanup target that should be met by the treatment (Cadotte et al. 2007; Toffoletto et al. 2005; Blanc et al. 2004; Volkwein et al. 1999), however, this requires that all compared technologies provide a certain minimum remediation efficiency. In addition to specifying the cleanup target, Toffoletto et al. (2005) and Godin et al. (2004) include a time frame for the remediation in their functional unit.

All studies have their focus on core components of the remediation project—on the on-site processing in terms of construction work and operational activities as well as ex situ activities such as soil treatment and/or deposition in

Table 1 Overview of reviewed literature, included technologies, and contaminants

Reference	Technology type		Contaminants	Pro- or retrospective aim
Beinat et al. (1997)	Not specified	Not specified	Not specified	Prospective
Diamond et al. (1999)	Ex situ	Excavation and disposal (S) Soil washing (S)	Generic examples	Both
	In situ	Soil vapor extraction (S)		
		In situ bioremediation (G)		
	Containment	Encapsulation (S)		
Page et al. (1999)	Ex situ	Excavation and disposal (S)	Lead	Retrospective
Volkwein et al. (1999)	Ex situ	Excavation and on-site ensured disposal (S) Excavation and decontamination (S)	PAH, mineral oil, chromium	Prospective
	Containment	Surface sealing with asphalt (S)		
ScanRail Consult et al. (2000)	Ex situ	Excavation and external biological treatment (S)	Chlorinated solvents, hydrocarbons	Both
	In situ	Biosparging (G)		
		Bioventilation (S)		
		Permeable reactive barrier (G)		
		Biological barrier (G)		
Vignes (2001)	Ex situ	Pump-and-treat (on-site vacuum steam stripping) (G)	TCP and total xylenes (G)	Prospective
		Pump-and-treat (on-site activated carbon treatment) (G) Excavation and thermal treatment (S)	Mix of organic contaminants (S)	
	In situ	Aerobic bioremediation (S)		
		Anaerobic bioremediation (S)		
	Containment	Cap and contain (S)		
Ribbenhed et al. (2002)	Ex situ	Thermal treatment (S) Bioslurry (S)	PAH, mercury, cadmium	Retrospective
		Soil washing (S)		
Blanc et al. (2004)	Ex situ	Excavation and off-site landfilling (S) Excavation and on-site containment (S)	Sulfur	Prospective
		Excavation and liming stabilization (S)		
		Excavation and bio-leaching (S)		
Godin et al. (2004)	Ex situ	Excavation and on-site secured disposal (S) Excavation and treatment (S)	Spent potlining landfill	Prospective
		Excavation and incineration (S)		
Toffoletto et al. (2005)	Ex situ	Excavation with on-site biopiles (S)	Diesel oil	Prospective
Bayer and Finkel (2006)	Ex situ In situ	Pump-and- treat (G) Permeable barrier (G)	PAH, Tar	Retrospective
Cadotte et al. (2007)	Ex situ	Pump-and-treat (G) Excavation with on-site biopiles (S)	Diesel oil	Prospective
	In situ	Natural attenuation (S, G)		
		Bioventing (S)		
		Chemical oxidation (G)		
		Biosparging (G)		
		Oil removal (NAPL)		
		Bioslurping (NAPL)		

The treated media is indicated after each technology as *S* soil, *G* groundwater, and *NAPL* nonaqueous phase liquid. *PAH* polyaromatic hydrocarbons, *TCP* 1,2,3-trichloropropane

Table 2 Functional units and time boundaries defined in LCA studies of soil/groundwater remediation

Reference	Functional unit	Time boundary
Beinat et al. (1997); Vignes (2001)	Not specified	Not specified
Diamond et al. (1999); Page et al. (1999)	Production of an equivalent amount of treated soil and groundwater (mass/volume)	25 years
Volkwein et al. (1999)	The ensemble of activities to achieve a certain risk level	Not specified
ScanRail Consult et al. (2000)	Not specified	Not specified
Ribbenhed et al. (2002)	1,000 kg of dry sediment into treatment	Not specified
Blanc et al. (2004)	A treatment of the site that allows environmental risks to be reduced to an acceptable level over the short term	Short term
Godin et al. (2004)	The management of 460,000 m^3 of waste mix and 200,000 m^3 of contaminated soil for a period of 50 years	50 years
Toffoletto et al. (2005)	Remediation during 2-year period of 8,000 m^3 of diesel contaminated soil to the Quebec B criterion	2 years
Bayer and Finkel (2006)	Control of a certain contaminated aquifer zone	30 years
Cadotte et al. (2007)	Remediation of a 375 m^3 diesel-contaminated site to the Quebec B criterion in soil (700 $mgkg^{-1}$) and to the detectable limit of C_{10}–C_{50} for potable, groundwater and surface water (0.1 mgL^{-1})	2–300 years depending on technology

landfill. In the latter case, landfill emissions are generally neglected due to lack of data (Ribbenhed et al. 2002; Page et al. 1999) or due to the short time frame of the LCA (Toffoletto et al. 2005), but transport of equipment, material, and soil to and from the site is included. As a common simplification, the manufacturing of hardware such as machinery, vehicles, and pumps used on-site is excluded in all studies except ScanRail Consult et al. (2000). Here, raw material use and emissions associated with the material constituents of these items are quantified according to use time relative to the total service life of the item. Materials for in situ and on-site installations, such as wells, barriers, activated carbon, etc. are generally included. For the ex situ soil treatment, all materials for construction of single-use on-site treatment facilities were inventoried (Toffoletto et al. 2005; Volkwein et al. 1999; Cadotte et al. 2007), whereas for permanent treatment facilities, only part of the impacts from construction were allocated to the remediation project based on its total capacity and assumed life time (Toffoletto et al. 2005; ScanRail Consult et al. 2000). Monitoring activities (person transport and laboratory analyses) are explicitly excluded in Bayer and Finkel (2006), Toffoletto et al. (2005), and Page et al. (1999), whereas Cadotte et al. (2007) includes transport of samples for analysis, but not the analysis itself due to its minor role. Two studies exclude activities that are identical for all compared remediation scenarios, i.e., power for pumping (Volkwein et al. 1999) and excavation work (Ribbenhed et al. 2002).

Some studies define a temporal boundary for the analysis (see Table 2). Diamond et al. (1999) argues that a time boundary of approximately 25 years is appropriate to capture long-term effects from no-action scenarios or waste storage. Cadotte et al. (2007) use a time boundary of up to 300 years depending on the time frame of the assessed remediation technologies. The long time frames are associated with a pump-and-treat scenario and a natural attenuation scenario. Toffoletto et al. (2005) and Blanc et al. (2004) define only short-term boundaries, whereas others do not specify the temporal boundary of their study. Although the REC methodology (Beinat et al. 1997) is based on life cycle principles, it neglects all material consumption and focuses only on energy use during the remediation project and the emissions related to this.

All studies focus on collection of specific data for the foreground processes taking place at the site and use average technology data for background processes such as production of electricity, diesel, steel, etc. Retrospective analyses largely base their data on information given in project completion reports from consultants and contractors. Prospective studies rely on site-specific modeling and dimensioning using simulation tools and analytic models.

2.2 Impact assessment

An overview of the environmental impacts covered in the applied impact assessment frameworks in the reviewed studies is presented in Table 3. Two of the reviewed studies (Vignes 2001; Blanc et al. 2004) do not carry out an impact assessment as such and are not included in the overview. Vignes (2001) includes characterization factors for 19 substances emitted to air, water, and soil, which are proportional to the inverse of the actual threshold value for a given compartment and converts all emissions to environmental impact units expressing how many times the

Table 3 Overview of impact categories used in the reviewed literature

Reference	Beinat et al. 1997	Diamond et al. 1999	Page et al. 1999	Volkwein et al. 1999	ScanRail Consult et al. 2000	Ribbenhed et al. 2002	Godin et al. 2004; Toffoletto et al. 2005	Bayer and Finkel 2006	Cadotte et al. 2007
LCIA method					EDIP97	USES-LCA[a]	EDIP97		TRACI
Environmental impacts									
Global warming		Q	x	x	x	x	x	x	x
Ozone depletion		Q			x		x		x
Photochemical ozone formation		Q		x	x	x	x	x	x
Acidification		Q		x	x	x	x	x	x
Nutrient enrichment		Q			x	x	x	x	x
Ecotoxicity		Q[b]			x	x[b]	x[b]		x[b]
Human toxicity		Q[b]	x	x	x	x[b]	x[b]	x	x[b]
Waste	x		x		x		x		
Surface water pollution	x								
Air pollution	x								
Land use	x[c]	Q[d]		x[c]					
Odor				x	Q				
Noise		Q		x	Q				
Other site-related impacts		Q[e]	x[f]						
Positive aspects	x[g]				x[h]				
Resource consumption									
Fossil energy	x	Q	x	x	x	x		x	
Scarce metals					x				
Clean groundwater	x	Q	x	x					
Clean soil/sand/gravel	x	Q	x		x				

Q categories that are used only for a qualitative impact assessment

[a] For eco- and human toxicity

[b] Includes both process-related (secondary) and site-related (primary) toxic impacts

[c] Land use due to remediation

[d] Land use due to landfilling

[e] Soil quality disturbance, heat damage, habitat alteration, effects on soil moisture, interrupted drainage, land stagnation, and human social disturbances

[f] Residual human toxicity burden

[g] Amount of cleaned soil and groundwater

[h] Prevented toxic impacts due to remediation calculated in a separate environmental benefit module

threshold value is exceeded. The summed environmental impact units then express the total environmental impact, and no impact categories are employed. The LCA conducted by Blanc et al. (2004) is intentionally terminated at the inventory level in order to avoid uncertainties and lacking consensus associated with impact assessment indicators. This choice is argued for by the fact that the focus is on resource productivity and not on environmental impacts.

Among the nine studies included in Table 3, all except one (Beinat et al. 1997) make use of some or all of the conventional impact categories: global warming, ozone depletion, photochemical ozone formation, acidification, nutrient enrichment (eutrophication), ecotoxicity, and human toxicity. However, especially the older studies seem to focus on few selected categories, which are then combined with additional categories important for the subject, e.g., land use (Volkwein et al. 1999; Diamond et al. 1999; Beinat et al. 1997), odor and/or noise (Volkwein et al. 1999; Diamond et al. 1999), and residual human toxicity burden (Page et al. 1999). Diamond and colleagues (1999) include all conventional impact categories, but only in a qualitative way. In addition to these conventional emission-related categories, Diamond et al. (1999) suggest a number of site-related impacts to be included in a general framework for life cycle management of contaminated site cleanup. These site-related impacts cover the physical change of the site due to the remedial activities, i.e., changes in soil quality parameters as well as damage to habitats and human social disturbances. More recent studies (ScanRail Consult et al. 2000; Godin et al. 2004; Toffoletto et al. 2005; Cadotte et al. 2007) employ established impact assessment methodologies at the midpoint level, i.e. Environmental design of industrial products (EDIP97; Wenzel et al. 1997) and The tool for the reduction and assessment of chemical and other environmental impacts (TRACI; Bare et al. 2003), and include most of the conventional impact categories.

In the REC methodology by Beinat et al. (1997), no actual impact assessment is conducted, since most impacts are given as their inventoried values, with one exception being “air pollution” which is derived directly from the energy use and expressed as population equivalents.

Apart from Blanc et al. (2004), consumption of finite resources is only reported in detail in ScanRail Consult et al. (2000), where four categories of energy resources and eight types of metals are inventoried and normalized. Other studies focus solely on the aggregated energy use given in energy units (MJ; Beinat et al. 1997, Volkwein et al. 1999, Ribbenhed et al. 2002) or as kilogram of crude oil equivalents (Bayer and Finkel 2006). Two studies focus on the use of local mineral resources (e.g., soil for backfilling of excavation pit) in their inventory (Beinat et al. 1997; ScanRail Consult et al. 2000). Another local or regional resource is groundwater use (e.g., for pump-and-treat), which is explicitly included in all earlier studies (Beinat et al. 1997; Page et al. 1999; Volkwein et al. 1999; Diamond et al. 1999).

The emission-related impact categories global warming, ozone depletion, photochemical ozone formation, acidification, and nutrient enrichment are treated rather similarly in the studies which consider them, reflecting the high degree of consensus on how to model these impacts. Larger differences are found in the applied models for quantification of ecotoxicity and human toxicity impacts. In all the studies, the toxicity models are at the midpoint level, i.e., the toxicity models include fate, exposure, and effect factors in the establishment of characterization factors, but do not extend the analysis to the endpoint where the potential damage is estimated, e.g., in terms of disability adjusted life years (DALY) or extinction of species.

The EDIP97 toxicity model is applied in three studies (Toffoletto et al. 2005; Godin et al. 2004; ScanRail Consult et al. 2000). In this model, the characterization factors for the human toxicity and ecotoxicity of a substance are calculated based on the key properties of the substance in terms of persistence, volatility, bioconcentration potential, redistribution in the environment, and (eco)toxicity. The factors express the volume of the compartment where the substance ends, which is needed to dilute the concentration to a level where no toxic effects are expected. EDIP97 models human toxicity via the compartments air, water, and soil. Ecotoxicity is separated into acute impacts in water, chronic impacts in water, and chronic impacts in soil (Wenzel et al. 1997).

Ribbenhed (2002) applies the USES-LCA toxicity model by Huijbregts et al. (2000), which is based on the Uniform system for evaluation of substances (USES 2.0). This model is a multimedia fate, exposure, and effect model including transfer between two geographical scales, continental and global. Four different ecotoxicity impact categories are included (freshwater aquatic ecotoxicity, freshwater sediment ecotoxicity, marine aquatic ecotoxicity, and marine sediment ecotoxicity) as well as one human toxicity impact category. Initial emission compartments are air, freshwater, seawater, industrial soil, and agricultural soil.

Cadotte et al. (2007) apply the TRACI impact assessment model developed by the US EPA (Bare et al. 2003). For evaluation of human toxicity, TRACI applies a regional multimedia fate model (CalTOX) coupled with a human exposure model including exposure via soils, ingestion of food and water, and inhalation. Human toxicity effects are divided into the three categories human health cancer effects, human health noncancer effects, and human health criteria-related effects. Potential ecotoxic effects as a consequence of emissions to air and surface water are combined to one impact category, ecotoxicity.

Bayer and Finkel (2006) expresses human toxicity in terms of arsenic (As) equivalents and do not include ecotoxicity in their impact assessment. Volkwein et al. (1999) use reciprocal values of so-called "Prüfwerte" as characterization factors for toxic releases to air, water, and soil taken from risk assessment guidelines. Toxicity via air is divided into two spatial scales, near-site emissions and remote emissions, respectively. Page et al. (1999) estimate the exposure concentrations of metals emitted to water using the Mackay level III multimedia model and compare these with no-effect doses to calculate toxicity ratios.

Normalization and weighting using Danish normalization references and weighting factors from EDIP97 (Wenzel et al. 1997) is done in ScanRail Consult et al. (2000), Godin et al. (2004), and Toffoletto et al. (2005). Bayer and Finkel (2006) and Cadotte et al. (2007) use German and Canadian normalization references, respectively. Godin et al. (2004) and Toffoletto et al. (2005) present the impact assessment result as single index values expressing the sum of the weighted impact scores, whereas Cadotte et al. (2007) present the sum of the normalized impact scores to a single index, i.e., inherently weigh all environmental impacts equally. Page et al. (1999), Ribbenhed et al. (2002), and Volkwein et al. (1999) do not apply normalization or weighting, but Volkwein et al. (1999) calculates so-called disadvantage factors where the impact scores in each category are normalized in relation to the technology with the lowest score. Beinat et al. (1997) uses an expert panel to weigh the included impacts.

2.3 Assessment of primary impacts

The primary impacts of a contaminated site have been defined as the site-specific impacts associated with the contamination itself as opposed to the secondary impacts related to the remediation activities (Volkwein et al. 1999). As different remediation techniques often will provide different levels of contaminant reduction resulting in different quantities of contamination left in the subsurface, this difference in remediation quality can be important to include in the life cycle assessment. In the reviewed literature, four overall approaches of dealing with this issue have been used: (1) Disregard quality differences or assume that all scenarios have equivalent cleanup efficiency (Bayer and Finkel 2006; Blanc et al. 2004). (2) Regard the total residual contaminant mass as a direct emission to soil, from where it is redistributed based on a multimedia steady state model involving relevant environmental compartments. This emission is either included in the overall impact assessment together with the secondary impacts (Diamond et al. 1999; Ribbenhed et al. 2002), reported separately as "primary impacts" (Toffoletto et al. 2005) or given its own impact category as the "residual human toxicity burden" defined by Page et al. (1999). It should be noted that Toffoletto et al. (2005) assumed that all the contaminant mass was emitted to the soil compartment with no redistribution. ScanRail Consult et al. (2000) expanded the existing EDIP toxicity model to include the compartment groundwater and used the model to evaluate the benefits in terms of saved toxicity compared to a no-action scenario (see further description in Section 3). (3) Set up a site-specific dynamic model for the contaminant fate at the site as done by Godin et al. (2004) and Cadotte et al. (2007) that used site-specific groundwater modeling to simulate the contaminant transport to a surface water receptor for different remediation scenarios. The primary impacts are in these studies solely accounted for in the surface water, whereas no primary impacts are accounted for in the soil or groundwater compartments. (4) Finally, some studies evaluated the primary impacts in a risk assessment separated from the LCA (Beinat et al. 1997; Volkwein et al. 1999).

2.4 Main results

On-site consumption of diesel and electricity is generally found to be among the most important cause of environmental impacts. Another major contributor is transport of excavated soil volumes in the case of off-site scenarios (Page et al. 1999; Godin et al. 2004; Ribbenhed et al. 2002). If the off-site scenario involved disposal of contaminated soil on land (landfilling), this was generally adding significantly to waste generation in the studies that included this (Godin et al. 2004; Page et al. 1999). Off-site treatment or disposal scenarios involving transport of excavated soil volumes generally concluded that transportation operations were important contributors to environmental impacts as well as bulk waste in the case of off-site disposal. The consumption of materials such as metals and plastics generally contributed only little to the assessment; however, both Ribbenhed (2002) and Bayer and Finkel (2006) mention steel production as a significant cause of impacts. Furthermore, the production of activated carbon for treatment of contaminated groundwater was found to contribute very significantly to the environmental burden in the studies where it was included (Bayer and Finkel 2006; Vignes 2001). In the two scenarios by Bayer and Finkel (2006), activated carbon is continuously used for water treatment and is responsible for more than 50% of the global warming potential. Other materials found to be important for the assessment is asphalt used for paving of soil treatment sites (Cadotte et al. 2007; Toffoletto et al. 2005).

Regarding ex situ remediation of diesel contaminated soil, Toffoletto et al. (2005) found that a permanent soil treatment facility was preferred to a single-use facility as long as the transportation distance to the permanent center

did not exceed 200 km. Blanc et al. (2004) concluded that on-site containment of sulfur-contaminated soil was preferred to off-site bioleaching or off-site landfilling. Cadotte et al. (2007) found that on-site biopile treatment of diesel-contaminated soil generated more environmental impacts than in situ soil treatment (natural attenuation and bioventing), and that the major cause for this was the asphalt paving of the treatment site.

Only few studies included in situ technologies for groundwater remediation. In Cadotte et al. (2007), in situ chemical oxidation is compared with in situ biosparging and pump-and-treat, which is an ex situ technology. In situ chemical oxidation was found to be the least preferable remediation option due to its significantly higher environmental impacts, which mainly stem from the oxidant production and transport.

In the studies where a normalization step was included in the impact assessment, most normalized impacts were in the same order of magnitude; however, in Cadotte et al. (2007), the normalized impact potentials for ecotoxicity (in water and soil) were generally three orders of magnitude higher than the second highest normalized impact and thus, dominated the impact assessment result. The reason for the high ecotoxicity is, according to Cadotte et al. (2007), unspecified oils stemming from diesel and electricity production.

In the studies evaluating both primary and secondary impacts (Godin et al. 2004; Toffoletto et al. 2005; Cadotte et al. 2007), it is generally found that primary impacts stemming from remaining contamination after remediation can be significant.

Whereas studies covered in this review all apply an attributional LCA approach on remediation of contaminated sites, Lesage et al. (2007a, b) recently applied a consequential LCA approach in their analysis of brownfield rehabilitation. In addition to assessment of primary and secondary impacts, they include so-called “ternary impacts” that account for the environmental impacts associated with greenfield habilitation, i.e., construction of new infrastructure as well as longer travel distances for people living on greenfields as opposed to rehabilitating a brownfield. The longer travel distances for residents on greenfields resulted in very extensive ternary impacts over a supposed 40-year occupation period, which significantly surpassed both primary and secondary impacts and made brownfield rehabilitation (by excavation) favorable over a risk minimization scenario, where the contamination was covered with 30 cm of clean soil.

3 Groundwater impacts in LCA

Life cycle assessment methodologies have frequently been developed for environmental assessment of industrial products, and impact assessment methodologies, therefore, often have well-developed models for air emissions, surface water emissions, and emissions to upper soil layer. However, when it comes to deeper soil layers and groundwater, these compartments have only gained little attention in established life cycle impact assessment methods. Recent applications of LCA within other sectors such as waste management (Christensen et al. 2007; Hellweg et al. 2005) and agricultural production (Geisler et al. 2004) have, however, brought attention to the importance of including groundwater impacts when conducting life cycle assessments.

In one of the studies reviewed here (ScanRail Consult et al. 2000), the environmental benefit of a remediation project is calculated and expressed in terms of saved toxicity via soil, air, surface water, and groundwater compared to a no-action scenario. The benefits are normalized to person equivalents (as applied in EDIP97). Since the EDIP toxicity model (as other LCA toxicity models) does not include groundwater as a separate compartment or considers emissions to deeper soil layers, the model is expanded in the study. A subdivision of the soil into surface soil (top 1 m), subsoil, and groundwater is made (see Fig. 1). Human toxicological exposure is assumed to occur via surface soil and groundwater (which is applied in sparsely treated form as drinking water for human consumption—in Denmark, the dominant source of drinking water is the deep groundwater) and ecotoxicological impacts through surface soil only. No toxic effects are assumed for the subsoil, but the long-term redistribution of contamination in this compartment is modeled based on steady state conditions between the upward diffusive flux (to surface soil) and the downward advective flux (to groundwater). Volatile contaminants are assumed to evaporate to the air and cause toxic impacts through both air and soil compartments. From the air compartment, 50% of the mass is assumed to end up in surface water following deposition. The groundwater compartment is so deep that it is considered as a final compartment from where no relocation takes place. In the study, characterization factors for human toxicity via groundwater are calculated based on the substance-specific Danish groundwater quality criteria (GWQC). In case of, e.g., PCE contamination reaching the groundwater, the characterization factor is determined according to Wenzel et al. (1997) as:

$$CF = \frac{1}{\mathrm{GWQC_{PCE}}} = \frac{1}{1\ \mu\mathrm{g/l}} = 1{,}000\ \mathrm{m^3/g}$$

An interpretation of this characterization factor is that for each gram of PCE removed from the groundwater, 1,000 m^3 of water is saved. EDIP97 has no normalization reference calculated for human toxicity via groundwater. As

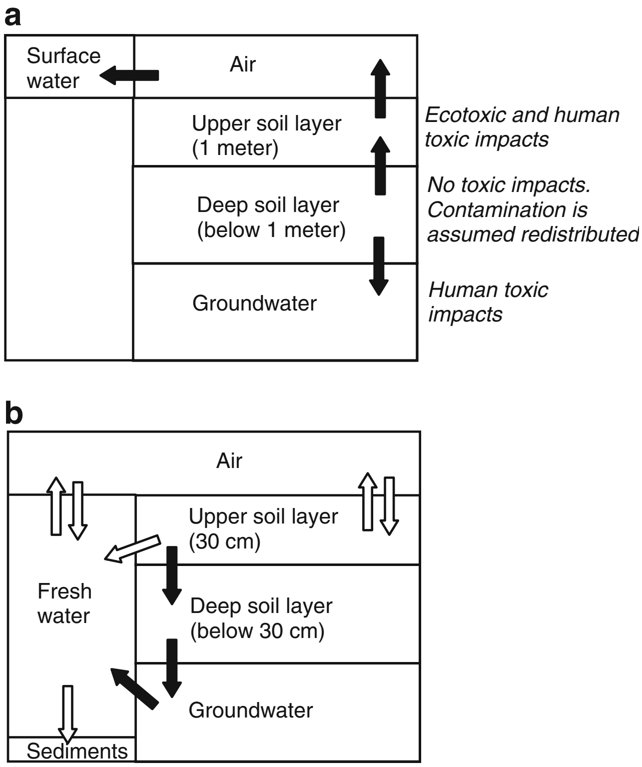

Fig. 1 **a** Illustration of subsurface compartments used for evaluating the environmental benefit of remediation by ScanRail Consult et al. (2000). **b** Model compartments used in Hellweg et al. (2005). The model includes transfer from soil to groundwater and from groundwater to surface water (*black arrows*)

a proxy, ScanRail Consult et al. (2000) use the total Danish groundwater reserve per person of 500,000 m^3 as a normalization reference.

Christensen et al. (2007) introduced a new impact category "spoiled groundwater resources" in their LCA model EASEWASTE (Environmental assessment of solid waste systems and technologies) to account for the nontoxic groundwater-contaminating effect of leached salts from landfills and waste residuals used in construction. Corresponding characterization factors are calculated as shown above using GWQC values for each salt. They express the volume of groundwater made unfit for drinking water due to salt contamination per mass unit of salt emitted to the groundwater compartment. The impact is normalized against the average annual Danish consumption of groundwater of approximately 140 m^3 per person. Furthermore, Christensen et al. (2007) included the impact category "stored ecotoxicity" (Hauschild et al. 2008) to account for the ecotoxicity potential stored in the landfilled waste after a set time frame. Characterization factors for this impact are calculated using the conventional EDIP97 ecotoxicity characterization factors assuming that of the remaining substances 50% eventually ends up in the soil and 50% in water.

Hellweg and colleagues (2005) provided a method for including deep soil layers and groundwater for leaching of heavy metals from landfill sites. They subdivided the subsurface environment into three subcompartments: upper soil, deep soil, and groundwater (see Fig. 1), and set up a site-dependent model describing the transport time to groundwater as function of infiltration rate, macropore flow, pH, organic content, and the distance to groundwater. The model was used to estimate concentration increase in upper soil, groundwater, and outflow to surface water at steady state and after certain time cuts.

4 Discussion

4.1 Functional unit and assessment of primary impacts

A key aspect of LCA is that the assessment compares different options for providing the same functional unit. When assessing different remedial options, this precondition is often not fulfilled since the remediation efficiency depends on the chosen technique. As long as a certain minimum cleanup criterion (set by the authorities) is fulfilled, different levels of residual contamination in the soil and groundwater can be considered acceptable, and all techniques meeting the cleanup criterion can be seen as providing a similar function. To define the functional unit based on the treated volume of soil and or groundwater[1], as many authors do, thus seems to be a suitable procedure. However, in order to be equally valid solutions for the decision maker, the expected remediation efficiency and project time frame should be within acceptable limits. These limits should preferably be stated in conjunction with the functional unit. Yet, the primary impacts can vary, e.g., as a consequence of a longer cleanup time which may result in larger primary impacts during remediation although the resulting reduction in contaminant concentrations is equal in the long term. By including the primary impacts in the LCA, the decision maker obtains a more fair comparison of the available remediation techniques.

An alternative to quantifying the primary impacts within the LCA framework is to evaluate them detached from the LCA as an element in an integrated assessment framework evaluating costs and benefits of a remediation activity. However, the advantage of quantifying the primary impacts within the LCA framework is that primary and secondary impacts are evaluated similarly and can be compared without needing to monetize them. The assessment of

[1] Note that in case of pump-and-treat systems, the groundwater volume may be the abstracted groundwater volume and not an aquifer volume.

primary impacts within the LCA framework requires that the benefit of complying with the regulatory standard (typically concentration based) must be converted to appropriate metrics for the applied impact assessment method, i.e., an avoided emission (mass based) if using the EDIP method.

4.2 Quantification of primary impacts

Several authors include both primary and secondary impacts in their assessment (Page et al. 1999; ScanRail Consult et al. 2000; Ribbenhed et al. 2002; Godin et al. 2004; Toffoletto et al. 2005; Cadotte et al. 2007) and the results confirm the relevance of doing this. Unfortunately, there is no consensus on how to quantify the primary impacts within the LCA framework. Some studies regarded the total contaminant mass as an emission to either soil or surface water; others used generic steady state multimedia models to forecast the long-term redistribution of the contaminant in the environment. Two of the reviewed studies (Godin et al. 2004; Cadotte et al. 2007) made a more site-specific assessment of the primary impacts applying groundwater modeling for evaluating the primary impact on the surface water receptor at risk with time.

As the primary impacts are indeed very site specific, this coincides with the generic nature of LCA and suggests that there is a need for more detailed impact assessment modeling on the site scale as is also being developed for leaching of metals from landfills (Hellweg et al. 2005). Another site-specific issue is the potential exposure to volatile organic contaminants (e.g., chlorinated solvents and benzene) in indoor air for residents at or near a contaminated site. This impact was not quantified in any of the studies dealing with this type of contaminants.

Quantification of primary impacts for a prospective LCA to be used in decision-support requires that the expected remediation efficiency (and thereby the residual contaminant mass) and remediation time can be predicted with some certainty. When it comes to in situ methods that rely on achieving certain conditions, e.g., to promote microbial degradation, the necessary treatment time to obtain a certain remediation goal is difficult to predict. This brings further uncertainty into the evaluation of the primary impacts. A reliable site-specific model describing the mass removal with time for the suggested remediation method is necessary to establish a likely range for the time frame, but requires that sufficient model data is available. This model should preferably include the generation of transformation products from naturally occurring degradation processes in the subsurface as these can impact the toxicity potential considerably (Gasser et al. 2007). An example is chlorinated solvents that are fairly persistent in subsurface environments, but can undergo anaerobic biodegradation. The parent compound is sequentially dechlorinated by specific degraders that have the ability to use the chlorinated compound as an electron acceptor in their respiration. As a result of this transformation process, there is a risk of accumulation of dichloroethylene as well as vinyl chloride of which the latter degradation product is more problematic than the parent compound and has a lower quality criteria value in groundwater due to is carcinogenic properties.

Since the toxic impacts have a large importance for the evaluation of primary impacts in LCA of site remediation, multimedia models with a higher level of complexity than the EDIP97 model could provide a more accurate characterization modeling of impacts associated with toxic substances. This would, however, also require more data. Generally, the uncertainty associated with estimation of toxic impacts is higher than for other impact categories in LCA due to the large uncertainty on the underlying substance data. Therefore, it is very relevant to conduct a sensitivity analysis for the most uncertain parameters in the evaluation of toxic primary impacts. However, the recent development of a consensus model (USEtox) for derivation of characterization factors for human- and ecotoxicity within the UNEP-SETAC Life Cycle Initiative has led to a better agreement between toxic impacts calculated with different impact assessment models (Rosenbaum et al. 2008). It should be noted, however, that an LCA of primary impacts on a contaminated site cannot replace existing environmental impact assessment methods for the identification of actual toxic impacts on the local scale as the LCA does not consider the sensitivity of the local environment.

4.3 Groundwater impacts and normalization

The reviewed studies mainly see contaminated sites as a soil contamination issue and do generally not address the potential impact of contaminating the groundwater. This may partially be due to the fact that groundwater is not included as an emission compartment in any of the applied LCIA toxicity models, which are thus not readily suitable for assessment of the primary impact in cases of contaminants that will potentially leach to the groundwater, e.g., chlorinated solvents. Another part of the explanation may reside in the fact that in many countries, groundwater is not an important drinking water source, or organic contaminants are removed (e.g., by activated carbon filtering) in the production of drinking water. The human exposure via direct use of groundwater will then be a minor issue in these countries, however, this is not a strong argument for neglecting this as other exposures through irrigation or discharge to surface water will be possible. The new EU Water Frame Directive emphasizes the importance of protecting groundwater bodies and the need to integrate the management of groundwater and surface water, recog-

nizing that they are linked. In this light, it seems reasonable to include groundwater as a separate compartment in LCA, which is interconnected to surface water receptors. This is consistent with Koehler (2008) that addresses the need for methodological solutions to properly account for freshwater use-related environmental impacts.

When introducing new impact categories for toxic effects in the groundwater regime, another important issue is the choice of appropriate normalization references in order to be able to compare the groundwater impacts with other impacts.

Furthermore, characterization factors used for assessing the primary impact could preferably be site specific, depending on the groundwater interests in the area. In this way, characterization factors for groundwater contamination in areas abandoned for drinking water supply could be lower than for groundwater contamination occurring within drinking water supply catchments or possible future water supply areas.

The normalization reference for human toxicity via groundwater suggested by ScanRail Consult et al. (2000) of 500,000 m^3 per person represents the maximum amount of groundwater that can be polluted per person, since the number represents the total existing groundwater resource per person. Thus, the normalized impact values are in fact minimum values. This volume, however, does not represent the volume of groundwater that can be exploited per person per year. The Geological Survey of Denmark and Greenland has evaluated the sustainable exploitable groundwater volume per person in Denmark to 190 m^3 per year, based on the Danish water resource model (Henriksen et al. 2008). This number represents the acceptable extraction rate if impact on groundwater quality and stream flow should not be compromised and even this figure would be a rather high normalization factor since it would assume that the total annual groundwater regeneration was contaminated due to man-made activities.

4.4 Long-term emissions and stored toxicity

The literature review revealed that emissions from landfilling of remediated soil were excluded due to lack of data or due to the long time frame of this emission. LCA methodology is in essence based on the principle of temporal justice. Emissions are aggregated over time, and no discounting of environmental impacts are carried out, i.e., an emission taking place in the far future is as important as one taking place now. Neglecting landfill emission strongly favors ex situ disposal scenarios over scenarios where an actual reduction in contaminant mass is achieved, e.g., in situ biodegradation. To ensure a fair comparison between remediation methods, the potential impact from landfilled soil should therefore be included in the assessment preferable divided into impacts occurring in the foreseeable future (t< 100 year) and a category representing the stored toxicity in the landfill (Hauschild et al. 2008).

The primary impacts from the contaminated site itself can also be regarded as a long-term emission. Contaminants like chlorinated solvents, which due to their low solubility have very extended leaching times, are likely to be potential sources of groundwater contamination for decades to centuries. Applying a dynamic transport and decay model for the emission of mass to the groundwater will allow for evaluation of primary impacts within a set time frame, e.g., 100 years. To account for contamination left in the soil after the time boundary of the analysis (e.g. 100 years), a category representing the stored human toxicity via groundwater could be introduced in analog to the stored ecotoxicity category in Hauschild et al. (2008).

5 Conclusions

The conducted review shows that life cycle assessment of site remediation options has been applied in a number of studies and that the tool is suitable for decision-support within this field. The vast majority of the reviewed studies consider ex situ treatment options, whereas in situ remediation technologies relevant for the frequently occurring DNAPL contaminated sites are only given little attention. The definition of the functional unit for conducting a comparative LCA of site remediation technologies is preferably done based on input rather than the output of treatment, since remedial efficiency varies. This entails a necessity of evaluating the primary impacts of residual contamination within the impact assessment.

The vast majority of the reviewed studies focus solely on contaminated sites as a soil contamination problem and do not address the potential impact of contaminating the groundwater (or related surface water bodies) when assessing the primary impacts of residual contamination. The inclusion of toxicity via groundwater as an impact category in LCA is an issue that requires future attention, since the groundwater compartment is not yet included and operational in established toxicity characterization models.

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